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Measurements and Standards for Nuclear Materials Safeguards

B. S. Carpenter and F. E. Jones

National Measurement Laboratory
U.S. Department of Commerce
National Bureau of Standards
Washington, DC 20234

March 1981

Annual Report, Fiscal Year 1980
(October 1, 1979 - September 30, 1980)

Prepared for
Division of Siting, Health and Safeguards Standards
Safeguards Standards Branch
Office of Standards Development
U.S. Nuclear Regulatory Commission
Under Contract No. AT(49-25)-9009

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U.S. DEPARTMENT OF COMMERCE, Malcolm Baldrige, *Secretary*
NATIONAL BUREAU OF STANDARDS, Ernest Ambler, *Director*

PREFACE

This report was prepared for the U.S. Nuclear Regulatory Commission, Office of Standards Development, under Interagency Agreement No. AT(49-25)-9009.

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ABSTRACT

This report is a review of the progress made during Fiscal Year 1980 (October 1, 1979, through September 30, 1980) of a long-term NBS program sponsored by the Nuclear Regulatory Commission (NRC) to upgrade national measurements and standards capability for nuclear materials safeguards. This is the last report to NRC for this program, since NRC has terminated support to NBS as of September 30, 1980. The NRC-sponsored program at NBS was a synergistic part of the overall NBS program in this area which is sponsored jointly by the NRC, the Department of Energy (DOE), and the Department of State through the International Safeguards Project Office (ISPO).

A summary of the progress for each of the five NRC-supported Tasks in the project is given. The first Task is concerned with the standardization of Non-Destructive Assay (NDA) methodology - work in this Task includes development of a series of low-enriched U_3O_8 NDA Gamma Spectroscopy NBS SRMs; the calibration of plutonium heat source reference materials for calorimetry; the establishment of a test facility for resonance neutron radiography (a possible NDA reference method) and the completion of experiments confirming the scientific validity of the method; effort toward using resonance neutron tomography (another possible NDA reference method) with reactor-produced neutrons for high-accuracy measurements and for the establishment of reference standards; and improvement of the accuracy of detectors to lead to more accurate measurement on the capture cross section of $^{238}UF_6$ gas.

The second Task involves the standardization of analytical chemical methodology - work in this Task includes: the development of a ^{233}U "spike" SRM for isotope dilution mass spectrometry; determination of the uranium content and isotopic composition of samples from the Safeguards Analytical Laboratory Evaluation (SALE) Program; and research to improve uranium mass spectrometry measurements.

The third Task involves the standardization of bulk measurement technology - the work in this Task includes: the standardization of tank volume measurements and the transfer of the technology to the field; the completion and laboratory testing of a prototype dynamic volume calibrator intended to make tank volume calibration more convenient, less time-consuming and less subject to operator error; the characterization of pressure transducers; the implementation of a Measurement Assurance Program (MAP) to improve in-facility weighing of large UF_6 cylinders; and field testing of a mobile flow prover for calibration of nuclear facility flow measurement systems at a nuclear facility.

The fourth Task applies mathematics and statistics to nuclear safeguards - the work in this Task includes: development and description in detail of a design for an improved measurement system and the associated statistical analysis of the resulting data; and recommendation of the appropriate statistical models for all currently used passive gamma, active gamma and passive neutron NDA measurement systems.

The fifth Task involves the development of a Users Guide to provide specific recommendations on how facilities should be designed to accommodate the necessary measurements to meet the SNM accountability goals of the safeguards program - the work in this Task includes the preparation of a review draft and preparation of the final version of the Users Guide for publication.

An adequate measurement and accounting system is necessary for the detection of and protection against unauthorized removal of special nuclear materials by persons having authorized access to facilities. The sensitivity of this type of detection depends directly on the uncertainties of measurement. The NBS program will assure the availability of the certified reference materials, reference measurement methods, reference data and quality assurance methodology for the adequate standardization of measurements for nuclear safeguards. Domestic and international dissemination is required.

The goal of the NBS program is

> To assure that measurement standards exist for the timely measurement of special nuclear material throughout the fuel cycles so that measurements can be performed at reasonable cost with accuracy sufficient for the safeguarding of nuclear material. These measurements of enriched uranium, plutonium, and related materials need to be made domestically by NRC and DOE inspectors, government facilities, and the industry. In addition, these measurements need to be made by other countries and the International Atomic Energy Agency (IAEA).

The overall NBS program is currently funded by NRC, DOE, and the Department of State through the International Safeguards Project Office (ISPO). NRC funding terminated on September 30, 1980. The program may be viewed as consisting of three related parts: (1) development of capability, including calibration standards, reference measurement methods, sampling schemes, statistical treatment of data, data generation; (2) dissemination mechanisms to transfer the standards and reference methods and data to the users; and (3) mechanisms to directly assist inspectors and the nuclear industry to ensure that their measurements are of sufficient accuracy.

In order to carry out the tasks assigned to NBS in a timely manner, NBS must continually assess the advancing needs for national and international measurement standards. NBS has received substantial guidance and input from NRC as to standards needs.

The NBS Measurements for Nuclear Safeguards Program is broadly organized into five Tasks: non-destructive assay; chemical and isotopic measurements; bulk measurmeents; statistics, sampling and error analysis; and the development of a users guide for design of facilities to accommodate accountability instrumentation.

Consideration for needed reference data is included in each of the first four Tasks. The research is being carried out in six line organizations at NBS: Center for Radiation Research, Center for Applied Mathematics, Center for Materials Science, Center for Thermodynamics and Molecular Science, Center for Analytical Chemistry, and Center for Mechanical Engineering and Process Technology. Researchers with backgrounds in analytical chemistry, mass and volume measurement, nuclear and radiation physics, thermodynamics, mechanics, etc., are part of the program team. NBS is also supplying a substantial amount of equipment, both previously existing and new, that is needed to carry out the program.

NRC supported an important portion of the total NBS program on measurements for nuclear safeguards in all of the above-mentioned Task areas.

TASK I: STANDARDIZATION OF NON-DESTRUCTIVE ASSAY (NDA) METHODOLOGY FOR NUCLEAR MATERIALS

The objective of this Task is to provide a common and accurate basis for the non-destructive assay of nuclear fuels and disseminate to industry, Government, other countries and the IAEA. This Task is divided into three components (Sub-Tasks): A - NDA Standard Reference Materials Research. The effort in this Sub-Task is directed toward the development of primary (facilities-independent) NBS-certified reference materials to be used for the calibration of NDA instruments. These SRMs will be used by others for the evaluation and development of NDA methods and for the production of NDA secondary or working reference materials. B - Reference Method Development. The effort is directed toward the development of reference methods to be used for the characterization of NDA reference materials.

Sub-Task A: NDA Reference Materials Research

A major portion of the effort in this Sub-Task has been on the development of a series of low-enriched U_3O_8 NDA Gamma Spectroscopy SRMs. This is a cooperative project involving NBL, LASL, NBS and Euratom Labs. NBS provided collaborative support to the Central Bureau for Nuclear Measurements (CBNM) on their design of a suitable aluminum container for the reference materials and helped to finalize the specifications for the U_3O_8 materials. Specifications were required for a number of parameters including: chemical assay, enrichment, chemical and isotopic homogeneity, and radioactive impurities.

Study of 20 pre-experiment samples of U_3O_8 prototype reference materials was completed to prepare the results for the Joint Collaborators' meeting scheduled in Geel, Belgium, 5-6 November 1979. The meeting consisted of representatives from all of the laboratories or organizations involved in this joint collaborative project: ESARDA, JRC-Geel, JRC-Ispra, Euratom Inspectorate, IAEA, KfK, NBL, LASL and NBS. The results, tabulated in Table 1, presented at the meeting consisted of:

- the homogeneity of each enrichment set (0.298%, 0.720% and 2.95%),
- the effects of varying packing density on a given enrichment (2.95%),
- the determination of ^{235}U enrichment,
- the problems associated with container design, and
- the problems associated with gamma spectroscopy, i.e., peak area evaluation, pulse pile-up, dead-time and collimation.

The results indicated that the material was homogeneous within each set and that there were no interfering impurities present. The variation in the packing density was found to have no effect on the enrichment measurements when the 186 keV line of ^{235}U was used. In all of the gamma spectroscopy data presented, consideration had to be given to, or corrections made for, pulse pile-up, dead-time, and variation in the window thickness of the cans. Finally, a new aluminum container design was approved (see Figure 1) which would protect the thin window from scratches.

A purchase order has been dispatched to British Nuclear Fuels, Ltd., to acquire the 4.5% enriched U_3O_8.

Table 1

^{235}U Enrichment Data on U_3O_8 NDA Test Samples

Sample	n	% Enrichment (Declared)	Measured Value
Depleted	5	0.298	0.2979 ± 0.0002
Natural	6	0.720	0.720 ± 0.001
Enriched	4	2.95	2.950 ± 0.002
Enriched with varying density	4	2.95	2.950 ± 0.003

The final design for the aluminum cans was given to the NBS Shops to mass produce 900 containers. Six of the first 20 cans produced were sent to Geel for tolerance evaluation by ultrasonic techniques. The Shops produced the cans in three phases: the inner plug, the outer can, and then the hollowing of the interior of the can. Figure 2 shows an array of outer cans and the inner plug being inserted into one of the outer cans. All 900 cans have been completed and shipped to CBNM-Geel for filling. At present, CBNM has taken 13 samples (∿1 g each) from the four batches of material (0.32%, 0.72%, 1.94% and 2.95% enrichment) that are being shipped to NBS for homogeneity and isotopic measurements. An additional 20-g sample from each batch is being shipped for chemical assay and for determining the ^{232}U concentration by gamma-ray spectrometry. CBNM has also begun canning the 0.32% U_3O_8 material and will continue in order of increasing enrichment.

Another major effort of this Sub-Task was devoted to studying the contribution of ^{238}U daughter products (^{234}Pa and ^{234m}Pa) to the 185.7-keV gamma-ray peak observed from low-enriched ^{235}U samples. The purpose of this study was to provide additional information in certifying the proposed low-enriched U_3O_8 NDA standards, as well as to aid in accurately measuring the ^{235}U. Since the protactinium gamma rays have energies at 185 ± 1 keV which cannot be resolved from the 185.7-keV gamma-ray line of ^{235}U, a small non-linear error is introduced in ^{235}U NDA enrichment measurements. According to the work of H. Ottmar at Kernforschungszentrum Karlsruhe GmbH, the contribution of Pa gamma rays to the 186-keV region is negligible (<0.1%) for enrichments at 1%, and is ∿0.5% for depleted uranium samples.

Our study was performed on previously prepared counting standards made from NBS uranium isotopic SRMs (see Table 2). These standards contained approximately 300 mg of U_3O_8 and were counted on a high-purity Ge detector for long periods of time such that errors due to counting were smaller than the effect being studied. Unfortunately, the sample sizes were too small to provide reasonable counting periods necessary for the accurate measurements needed.

In order to increase the accuracy of the measurement and reduce the counting time needed, larger sample sizes (∿11.5 g) were prepared. Gamma-ray spectrometry has been performed on 11.5-gram samples whose ^{235}U isotopic abundances have ranged from 0.0004% to 0.7198%. This isotopic range was chosen because the contribution from ^{234}Pa and ^{234m}Pa is significant.

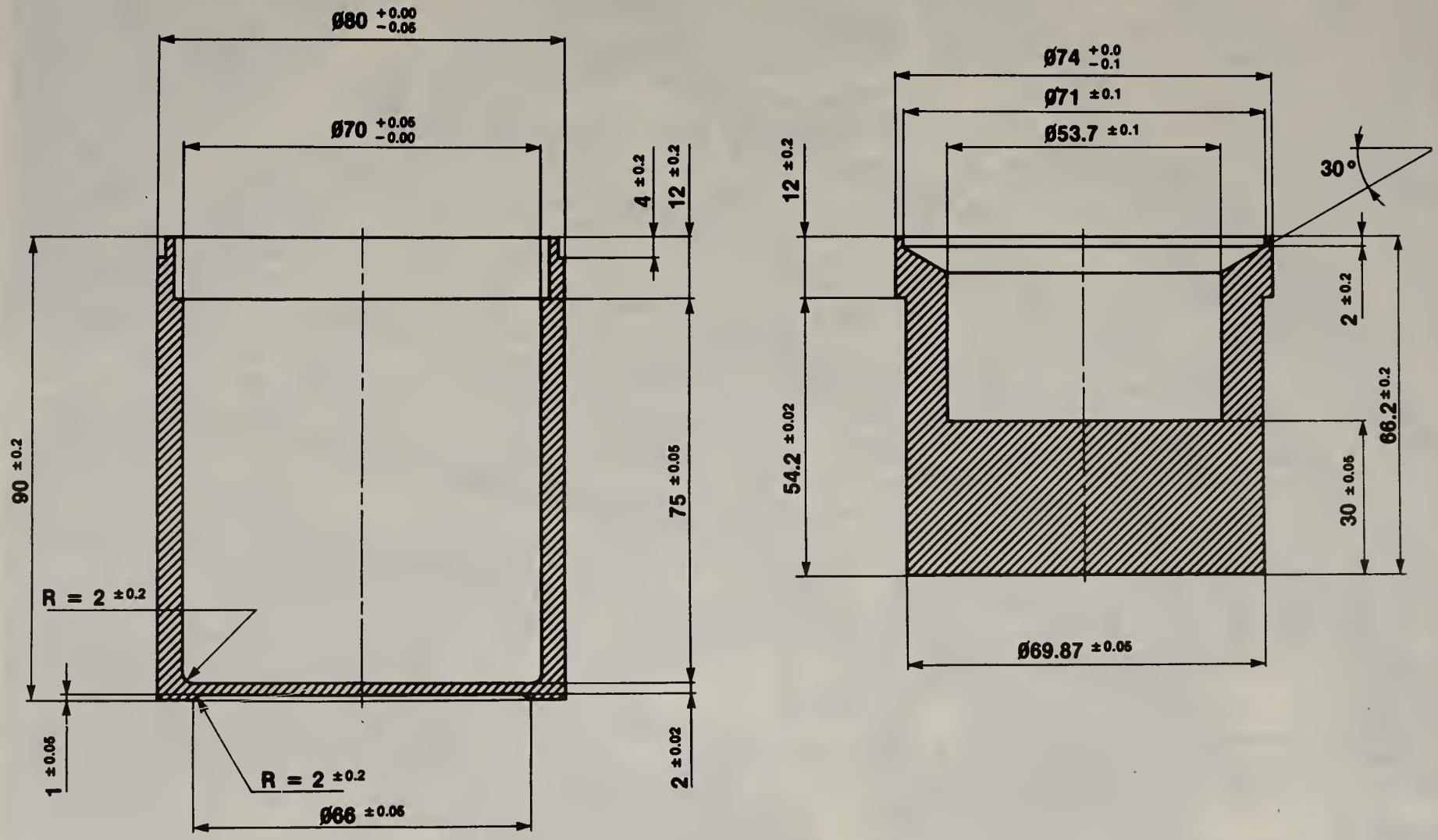


Figure 1. New Aluminum Container Design Drawings

Figure 2. An Array of Outer Cans and the Inner Plug Being Inserted Into One of the Outer Cans

Table 2

Previously Prepared Counting Standards
Made From NBS Uranium Isotopic SRMs

SRM	% ^{235}U	% ^{238}U
U-0002	0.01755	99.9823
U-005	0.4895	99.504
U-010	1.0037	98.984
U-015	1.5323	98.443
U-020	2.038	97.933
U-030	3.046	96.915
U-050	5.010	94.915
950a	0.7198	99.2711
U-238	0.004	99.995

In addition to the gamma-ray spectrometry, these samples are to undergo chemical separation, since secular equilibrium exists. The uranium is separated from its daughter decay products of thorium and protactinium. After separation, the two fractions (uranium and thorium-protactinium) are then measured by gamma-ray spectrometry. By measuring the uranium fraction, there is an indication of the 186-keV gamma-ray line build-up from protactinium, while the thorium-protactinium fraction provides quantitative information on the actual amount of interference being contributed.

The investigators for this Sub-Task are B. S. Carpenter and T. Mitlehner.

NDA Calibration Methodology

Major steps were taken in FY 80 to realize the NBS goal of providing traceability to national standards for calorimetric measurements made in the industrial assay of plutonium-bearing solids. These included (1) further testing of a Mound-designed heat-flow calorimeter, together with an automated data-acquisition system (this included power measurements on two Mound-fabricated, encapsulated plutonium heat standards); (2) design and start of construction of an NBS heat-flow calorimeter; and (3) preparation of an NBS "General Reference Material" certificate to accompany Mound-calibrated plutonium heat standards.

The twin-bridge, heat-flow calorimeter obtained previously on loan from the Mound facility was tested in measuring the total decay power of two calibrated plutonium heat sources (nominal powers 0.25 W and 4.0 W, respectively). The sources were loaded automatically and calorimeter data were recorded with an automated data acquisition system built around a Hewlett-Packard 9835* desktop computer. The NBS power data for both sources had a range of 20 ppm and a standard deviation of about 10 ppm. They agreed with the Mound predicted powers for these sources within the combined measurement uncertainties of NBS and Mound.

*Identification of commercial equipment, instruments, or materials does not imply recommendation or endorsement by the National Bureau of Standards.

Design specifications for an NBS twin-bridge, heat-flow calorimeter were developed, using our experience with the Mound calorimeter as a guide. While the gross physical features and operating principles of the NBS calorimeter follow the Mound design, some changes in construction materials (as use of stainless steel for all submarine parts and use of O-rings instead of gasket seals) were used to improve the reliability. In addition, the calorimeter was designed to be symmetrical, so that either of its two sensing chambers could be used as a reference chamber. The main calorimeter body has been completed, and we are presently constructing the thermal sensing elements, including heaters and resistance thermometers.

In order to establish traceability to national standards for calorimetric heat-flow measurements in assaying radioactive materials, we are proposing a service similar to the one offered by NBS to the radiopharmaceutical industry: essentially a Measurement Assurance Program (MAP) in which traceability to national standards is established through a programmed exchange of measurement artifacts between major suppliers (in this instance, of calibrated radioactive heat sources) and NBS. As a first step in this direction, we have carried out power measurements on three Mound-calibrated heat standards in an NBS ice calorimeter, and have determined that the NBS- and Mound-measured powers are in agreement within their combined uncertainties for all three standards. We are therefore, together with the Office of Standard Reference Materials, preparing a "General Reference Material" certificate (GM-10) that documents these NBS/Mound intercomparisons. Mound will be authorized to distribute this certificate (together with its own calibration certificate and data) along with each newly-calibrated source it provides. A copy of the NBS certificate for GM-10 appears in Figure 3. As soon as the NBS heat-flow calorimeter is operational, a formal MAP between Mound and NBS, covering calibrated plutonium heat standards, will be started.

David Ditmars, the staff member whose measurements were used in the preparation of Special Reference Material Report, GM-10, "Encapsulated Plutonium Heat Source", also serves as a member of INMM 8.4, "Calibration Techniques in the Calorimetric Assay of Pu-Bearing Solids", and the NBS Pu Steering Committee.

The investigators for this portion of the Sub-Task are D. A. Ditmars and J. Colwell.

Sub-Task B: Reference Method Development

Resonance Neutron Radiography

Work continued on improving experimental appratus and methods of data reduction for resonance neutron radiography as an NDA technique. Having established the scientific validity of the method, we have been actively engaged in reporting our results at several conferences. Our preliminary tests to develop the resonance neutron radiography technique involved the nondestructive examination of a defective silver braze. These results have been finalized and prepared for publication.

Experimental gas-filled position-sensitive proportional counters (PSPCs) are used in the development of the resonance neutron radiography technique. For this application, "area" PSPCs having submillimeter spatial resolution for neutrons in the epithermal energy range are needed. Through the NBS collaboration with Oak Ridge National Laboratory (ORNL), a "linear" PSPC was built having

Figure 3.

Special Reference Material Report

GM 10

Encapsulated Plutonium Heat Source

Introduction

The enclosed encapsulated plutonium heat source provides a convenient, measured standard for calorimeter calibration [1] in the calorimetric assay of plutonium-bearing solids. This report summarizes the NBS measurements made on this reference material. The reference is produced and certified by the Mound Facility*, Monsanto Research Corporation, Miamisburg, Ohio.

Description of Source

This heat source, fabricated and calibrated at the Mound Facility, consists of a doubly encapsulated sample of plutonium-oxide. The sealing of both inner and outer capsules has been carried out using documented and reproducible welding procedures. Nondestructive testing (radiography and He leak tests) has been conducted to ensure the integrity of each capsule. Since the active materials are largely alpha-particle emitters, helium gas will accumulate within the inner capsule. The capsules have a conservative design life of five years. The lack of long-term compatibility and creep data for the materials of construction precludes the assignment of longer working lifetimes to the sources at this time. Therefore, it is strongly recommended that this source be returned three years from the date of receipt to the Mound Facility for reevaluation and recertification. This source should be handled with the normal precautions for alpha-particle emitters and stored in the container provided when not in use.

Certification

This source has been calibrated at the Mound Facility using Mound heat-flow calorimeters and electrical standards traceable to the National Bureau of Standards. A separate certificate issued by the Mound Facility accompanies this reference material. Selected Mound plutonium heat sources similar to the one accompanying the Mound certificate have been independently measured in a precision Bunsen ice calorimeter at NBS. The calibration results accompanying that certificate are in the form of computer printout and microfiche giving the decay power calculated from these measurements, day-by-day, for a three-year period after the calibration. Also included are the physical parameters of the source including materials of construction and measured radiation dose rates.

*Mound Facility is operated by Monsanto Research Corporation for the U.S. Department of Energy under Contract No. DE-AC04-76-DP00053.

Figure 3 (cont'd.)

NBS Measurements on Mound-Encapsulated Sources

Each measurement in a precision Bunsen ice calorimeter reflects the average (unweighted) of at least 10 independent determinations of heat flux (power) for a single source. Three such measurements were carried out on Mound sources of nominal power, 0.23W, 1.5W, and 1.0W, and are summarized below. The first two are documented extensively in [2].

	Mound Source Designation		
	0.23WB	1.5WB	1.0WK
Mound Predicted Power[a][W]	0.22544	1.44942	0.97027
NBS-Measured Power[a] [W]	0.22517	1.44936	0.96996
Number of NBS Measurements	16	19	11
s_m[b] [W]	0.00005	0.00004	0.00034
Estimated NBS Overall Uncertainty[c] [W]	0.0003	0.0007	0.0014

[a] Individual data for each source calculated for a single time mid-way through the measurements series for that source.

[b] s_m = Computed standard deviation of the mean.

[c] Sum of maximum conceivable systematic errors and 99% confidence limits for the mean.

At NBS measurements were carried out by D. Ditmars of the Chemical Thermodynamics Division and at Mound Facility by K. C. Jordan, Senior Research Specialist.

Packaging and reshipment of this source to the Mound Facility for testing and recalibration must follow all applicable Department of Transportation, U.S. Department of Energy and/or U.S. Nuclear Regulatory Commission regulations. All shipments should be addressed to:

C. L. Fellers, Group Leader
Safeguards Research and Development
Monsanto Research Corporation
Mound Facility
P.O. Box 32
Miamisburg, Ohio 45342

[1] ANSI Standard N15.22-1975, "Calibration Techniques for the Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Materials Control", available from American National Standards Institute, 1430 Broadway, New York, New York 10018.

[2] D. Ditmars, Intl. J. Appl. Radiat. Isotopes 27, 469 (1976).

a spatial resolution of about 1 mm. High spatial resolution area PSPCs have now been built to replace the linear PSPC. These area PSPCs contain 3 atm. ^3He, 11.5 atm. Xe, and 0.5 atm. CO_2. The sensitive area is 50 x 50 mm^2, and the spatial resolution is 1.5 x 1.5 mm^2. Work continued on improving this design and implementing the new detector for use on the linac. Preliminary runs have verified resolution of about 1 mm. The counter gas has been modified to increase the amount of ^3He gas by a factor of 5 to increase the detector sensitivity. Modifications have been made in the voltage divider chain to allow operation at 20% lower bias potential to avoid gas breakdown. Tests of the effects of these changes are now underway.

A sample of spent fuel is now being acquired to determine the radiographic technique sensitivity to the fission products and plutonium isotopes present. Work on unirradiated fuel pellets was reported in the Annual Report for FY 1979 in a paper presented at the American Nuclear Society Topical Meeting, November 26-30, 1979, Kiawah Island, SC, published in the Proceedings for the meeting (Safeguards Reference Measurement System Utilizing Resonance Neutron Radiography, by R. A. Schrack, J. W. Behrens, C. D. Bowman, and A. D. Carlson).

The neutron radiography detector system has been used in a pin-hole camera system to image the distribution of neutrons from a spatially extended source (see Figure 4). The source in this case was the neutron-producing target used with the NBS electron linac. Adjacent to the target was a 1/16"-thick cadmium sheet with a "V" cutout. In the experiment the low-energy ($E_n < 0.3$ eV) neutrons emerging from the V were imaged. To image the V we placed a 1/16"-thick cadmium sheet, containing a 2-mm diameter hole in the center, at a flight distance of 4 meters from the neutron target. A one-dimensional PSPC was placed horizontally at 4.5 meters and could be remotely positioned in the vertical direction. The PSPC, designed and built in collaboration with the Instrumentation and Control Division of ORNL, had a sensitive length of 50 mm and a spatial resolution of $\sim$ 1.2 mm. The results of this measurement were presented at the June 1980 ANS meeting in Las Vegas, Nevada.

In Figure 5 the results are shown. A total of nine horizontal slices were required to image the V. The circle in Figure 5 represents the field-of-view imposed by our flight path collimation. The pixel size and the resolution are also indicated.

Calculations were carried out to determine the degree of non-homogeneity of an absorber that can exist within a pixel without degradation of assay accuracy. The ability to tolerate greater pixel inhomogeneity allows quicker measurements because larger areas can be covered in a single measurement. The computer calculations have shown that inhomogeneities in sample thickness that would cause large errors in assay of constituents can be compensated for and the error reduced by a factor of about 20 by shape analysis of the resonance line in the absorption spectrum. With this philosophy in mind, two samples of spent fuel have been measured treating the whole 1-cm diameter slice of fuel as one analysis area. Analysis of the data is now underway, but a cursory examination shows quite obvious differences in isotopic composition between the two samples. The two samples are from the same fuel rod. One sample is taken from the end of the rod where the neutron flux is relatively low, and one cut is taken from the center of the rod where the neutron flux is greater. It is clear that the transuranic isotope buildup is greater in the center cut and, conversely, the depletion of the ^{235}U is less on the end cut. This technique should be a valuable tool in spent fuel analysis for the safeguards program to determine reactor history and fuel rod composition.

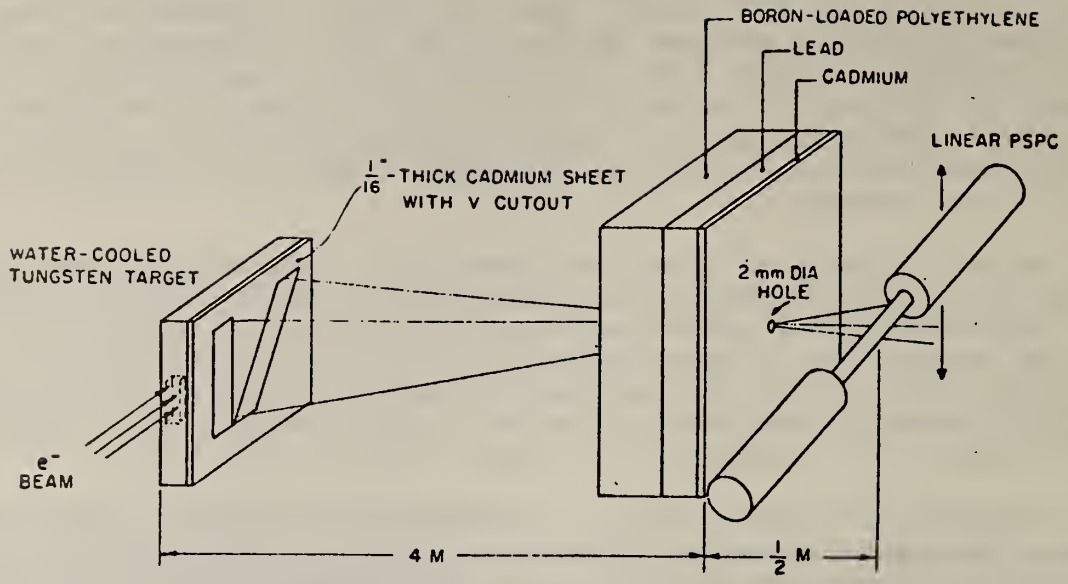


Fig. 4. Experimental arrangement for neutron pin-hole camera

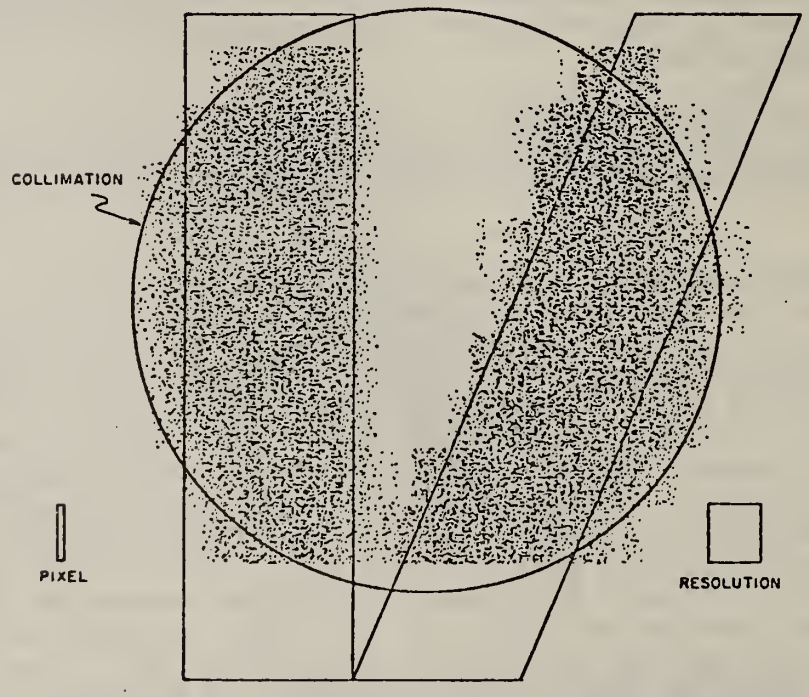


Fig. 5. Pin-hole camera image of the "V" obtained with low-energy neutrons

A different type of two-dimensional detector has been developed in cooperation with Surface Science Corporation. A small detector about 2.5 cm in diameter, having a resolution of about 0.1 mm, has been developed that utilizes a multichannel plate electron amplifier. Plans for testing the device are now underway.

The following papers were written during this fiscal year:

J. W. Behrens, R. A. Schrack, A. D. Carlson, and C. D. Bowman, "Resonance Neutron Radiography for Nondestructive Evaluation and Assay Applications", presented at Conf. on Nuclear Cross Sections for Technology, Univ. of Tenn., Knoxville, TN, Oct. 22-26, 1979, Proceedings published as NBS Special Publication 594, September 1980.

R. A. Schrack, J. W. Behrens, C. D. Bowman, and A. D. Carlson, "Safeguards Reference Measurement System Utilizing Resonance Neutron Radiography", presented at Conf. on Measurement Techniques for Safeguards and Materials Control, Kiawah Island, SC, Nov. 26-30, 1979, NBS Special Publication 582, June 1980.

J. W. Behrens, R. A. Schrack, and C. D. Bowman, "Nondestructive Examination of a Defective Silver Braze Using Resonance-Neutron Radiography", Nuclear Technology 51, No. 1, pp. 78-82, November 1980.

The investigators for this portion of the Sub-Task are R. A. Schrack, J. W. Behrens, A. D. Carlson, C. D. Bowman and R. G. Johnson.

Resonance Neutron Tomography

The effort on the resonance neutron tomography portion of this Sub-Task during FY 1980 was devoted to two areas: (1) the continuing development of software programming and the library data set; and (2) the finalizing of the design of the reactor-based resonance neutron safeguards experimental system and the making of provisions for its use. The major effort was concentrated on the second area.

In the first area, a mathematical analysis has been conducted and a computer code written for solving multiple non-linear equations relating fission detector response to fissile sample thickness. The cross-section data library has been further upgraded; this has helped to improve the sensitivity for the self-indication effect.

The detail design of the reactor-based resonance neutron safeguard experimental system was completed in April and has been approved by the NBS Reactor Hazards Evaluation Committee. The NBS Shops Division is responsible for the construction of the system and the letting of bids to private contractors.

Three U-235 fuel pellets of different enrichments were acquired and are being fabricated into 1-mm thick discs for use in the initial resonance neutron transmission experiments. An experiment is scheduled in early September to utilize these discs to verify and implement our theoretical predictions.

In addition, glovebox provisions are being made to the SRM Shop to permit the handling of SRM U_3O_8 powders of various enrichments for the fabrication of transmission cells.

In order to verify the resonance neutron self-indication effect with the NBS reactor facility, an experiment was conducted to quantitatively determine the U-235 and U-238 contents in three nuclear fuel pellets supplied by General Electric Wilmington Manufacturing Department. The known information is in Table 3.

Table 3

Known Characteristics of Three Nuclear Fuel Pellets
Supplied by General Electric Wilmington Manufacturing Department

Isotopic U-235 enrichment %	% U	Density	Length	Diameter
3.9572	88.15	95.9243	0.432"	0.410"
2.5174	88.168	96.56	0.4395"	0.410"
1.194	88.14	95.52	0.434"	0.410"

A horizontal neutron beam of 1/4" (0.635 cm)-diameter was extracted from the NBSR core on port BT-7. The thermal neutron portion of the beam was removed by using a 0.04" (0.102 cm)-thick Cd sheet as filter. The filtered beam was then transmitted through the fuel pellet and detected by NBS double fission ionization chamber detectors [Grundl, J. A., Gilliam, D. M., Dudey, N. D., and Dopek, R. J., Nuclear Technology 25, 237 (1975)].

The experimental setup and fuel pellet holder are shown in Figure 6. The holder was made with a lucite block approximately 15 cm long, 5 cm wide and 1 cm thick. Four holes were drilled to accommodate the three fuel pellets and a blank reference hole. The three fuel pellets, along with the blank hole, were scanned by the neutron beam and detected with U-235, Pu-239 double fission chamber detectors and then with U-235, U-233 detectors. The reason for using Pu-239 and U-233 detectors was that the fission cross section for U-238 was very small. A count rate of $\gtrsim$ 1000 cps was obtained using very thin fission foil in all detectors, thus no corrections were made for detector dead-time, self-absorptions, and background noise. A total count of $\gtrsim$ 50,000 was accumulated for each data point.

Table 4 lists the measured U-235 and U-238 contents in grams in the third pellet as compared to the known quantities.

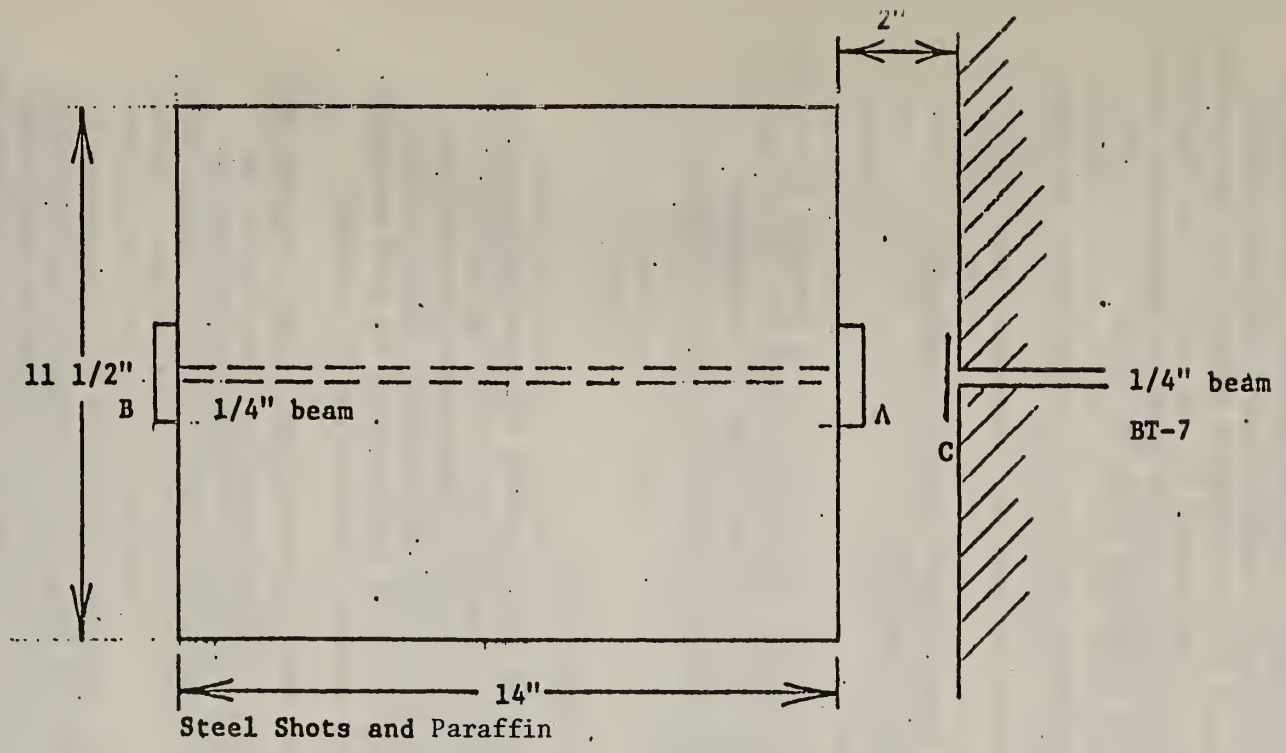
Table 4

Measured U-235 and U-238 Contents, in Grams,
in the Third Pellet as Compared
to the Known Quantities

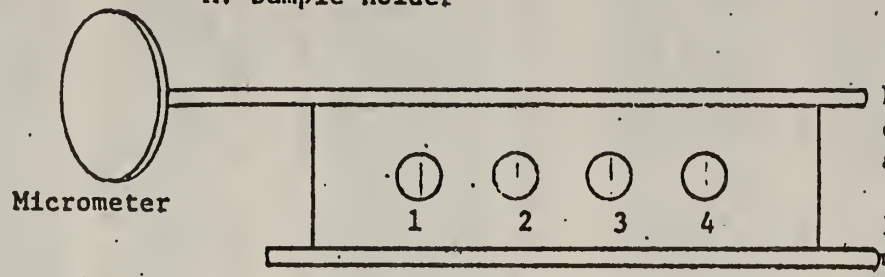
	Known quantities*	Measured with U-235, Pu-239 detectors**	Measured with U-233, U-235 detectors**
U-235	0.2174	0.185 $\pm$ 0.03	0.193 $\pm$ 0.03
U-238	5.2764	5.581 $\pm$ 0.05	5.476 $\pm$ 0.49

*Corrected for 1/4" neutron beam size.
**Errors were estimated second order effects.

The measured U-235 content in the third pellet is about 11% of the known U-235 quantity when the U-233 and U-235 detectors set are used, and about 15% with the U-235, Pu-239 set. The accuracies for the measured U-238 content in the third pellet are 4% and 6%, respectively, for the two sets of detectors.



A: Sample Holder



Lucite block 15 cm x 5 cm x 1.07 cm diameter of the four holes 1.04 cm top and bottom of lucite are covered by .025' Al sheets. The position of the lucite will be controlled by a micrometer. Holes #2, #3, and #4 will contain the pellets.

B: Fission Detectors
NBS double chamber fission detectors
U-233, U-235 and Pu-239 3 mg/cm^2 thick

C: Cd Sheet .102 cm thick

Figure 6. Neutron Telescope and Fuel Pellet Holder Assembly

The fact that all neutrons with energies below 0.5 eV were filtered out in the beam caused the Pu-239 detector to perform less satisfactorily relative to U-233 and U-235 detectors, since 60% of the resonance strength for Pu-239 lies in the 0.3 eV region. Nevertheless, the results indicate that a reasonably accurate estimation of U-235 and U-238 contents can be obtained in a very short time ($\lesssim$ 1 min.) with the self-indication method with counting statistics of 0.1%.

In summary, it would require approximately one day to characterize a nuclear waste container approximately the size of a gallon paint can for use as a reference material or to verify field methods. This conclusion assumes that it would require 110,000 transmission measurements with a resolution of 4 mm and 3% counting statistics. The assumption is made that the container will be scanned at 64 points in a plane orthogonal to its axis. The container will be rotated in two-degree increments following each orthogonal 64-point scan. Following each complete 180° rotation cycle, the container will be stepped axially and the process repeated for a total of 20. The amount of time needed for the entire interrogation, together with resolution and counting statistics, can be greatly improved by incorporating a linear series of detectors and a reactor power of 20 MW.

The investigators for this portion of the Sub-Task are D. A. Garrett, Y. T. Cheng, and M. Ganoczy.

TASK II: STANDARDIZATION OF DESTRUCTIVE ANALYTICAL CHEMISTRY METHODOLOGY FOR NUCLEAR MATERIALS

The goal of the Destructive Analytical Chemistry Task is to provide standardization for wet chemical and mass spectrometric methodology used for the assay of nuclear materials. This Task is divided into two Sub-Tasks: A - Standard Reference Material (SRM) Research and Reference Methodology; and B - Support of Measurement Assurance Programs (MAPs).

Sub-Task A: SRM Research and Reference Methodology

The goal of this Sub-Task is certification of a ^{233}U "spike" SRM for both uranium elemental concentration and isotopic composition. This material is intended as a spike for mass spectrometric determination of the isotopic and elemental composition of uranium in the full range of materials found in the nuclear fuel cycle. With this material, a single analysis can be used to obtain both the elemental concentration and the isotopic composition of uranium.

Approximately 400 ampoules of the ^{233}U "spike", SRM 995, were prepared containing 5 mg U in 10 mL of HNO_3 (1 + 19). Before ampouling, the uranium was purified by anion exchange chromatography to remove ^{229}Th and other daughter products. All analytical measurements and an extensive statistical evaluation of the SRM were completed.

The concentration of uranium in the ampoules of the SRM were determined by thermal ionization isotope dilution mass spectrometry following a sampling plan supplied by the NBS Center for Applied Mathematics: eight ampoules were chosen for use in the assay of the SRM. Separate aliquots from each of the ampoules were spiked with known amounts of ^{235}U (SRM 993) and uranium metal (SRM 960, assay standard). In addition, eight aliquots of SRM 993 were spiked with SRM 960 as a cross-check on the assay of these materials. Thus, all three SRMs have been assayed against each other.

The statistical design of the experiment allowed for the assessment of the following parameters: ampoule-to-ampoule variations among the ^{233}U samples, sample-to-sample variations among the ^{235}U and natural uranium metal spikes, inconsistency in the concentration previously assigned to the ^{235}U spike SRM, and finally the measurement error in the concentration of the ^{233}U spike SRM. No ampoule-to-ampoule or sample-to-sample variations were detected from the statistical evaluation of the data. The concentration of ^{233}U spike (SRM 995) was found to be 489.93 μg U/g with an uncertainty at the 99% confidence interval of ±0.037%. This uncertainty can be divided into a 0.011% random error component and a ±0.026% possible systematic error. The isotopic composition of SRM 995 was also determined mass spectrometrically using both Faraday cage and ion counting detection systems. The ^{233}U content of the SRM was found to be 99.9245 ± 0.0006 atom percent.

One member of the NBS staff visited the New Brunswick Laboratory, Argonne, Illinois, for the purpose of observing their methods for small sample assays of uranium. It is a goal of the NBS laboratory to try to determine uranium (assay) at a very small sample size while retaining as much of the accuracy as possible. Certification of the absolute isotopic composition of uranium SRMs requires the mixing of pure separated isotopes to produce calibration standards of known isotopic composition. High accuracy assay techniques for small samples are needed to minimize the irreversible use of high purity uranium separated isotopes. The NBL laboratories have been routinely analyzing ∿ 50 mg samples of uranium with a precision of ±0.05% or better. Preliminary studies at NBS indicate that the titration itself may yield precisions of ±0.01% at a sample level of 50 mg.

A visit was made also to the Central Bureau of Nuclear Measurements in Geel, Belgium, where experiments on the use of the Davies-Gray titration method in the assay of small uranium samples are being carried out. In this laboratory, 250-300 mg samples were being titrated to an automated end point with a precision of 0.01%. A contributing factor to this improved precision was a higher purity of starting material.

With the arrival of a printing digital pH meter, some efforts have been made to scale down the Davies-Gray titration. At a level corresponding to 100 mg of U metal, it is possible to titrate dichromate manually to a precision of better than ±0.01%. The dichromate titration itself may be useful at a level equivalent to 10 mg uranium. Future experiments will center around miniaturizing the Davies-Gray chemistry to these levels while retaining as much precision as possible.

Sub-Task B: Support of Measurement Assurance Programs

High precision and high accuracy measurements are made at NBS to support MAPs by providing the "true value" for uranium elemental concentration and isotopic composition of reference samples distributed in selected national and international evaluation programs.

Atomic weights of depleted and enriched SALE samples were computed based upon the isotopic analysis of these materials completed by the mass spectrometry group. Using the calculated value for the depleted SALE sample, the correct assay values for the depleted SALE samples were calculated. The results previously reported were based upon a natural uranium atomic weight and were subject to revision pending the true atomic weight calculation.

The assay of the enriched SALE samples was determined and reported based upon the NBS calculated atomic weights. During these assays, a major problem was found with contamination of the sample from beakers previously used for other uranium assays. A procedure was developed to positively control the uranium blank problem.

The investigators for this Task are J. D. Fassett, E. L. Garner, J. W. Gramlich, H. M. Kingston, L. A. Machlan and J. R. Moody.

TASK III: STANDARDIZATION OF BULK MEASUREMENT METHODOLOGY IN NUCLEAR FUEL CYCLE PLANTS

This Task provides standardization for bulk measurements (mass, volume, density, flow, pressure and temperature) of nuclear fuels. Bulk measurements play a very important role in the accountability of SNM and in the safeguarding of SNM against diversion. The standardization provided by this Task will assist inspectors and licensees in implementing efficient measurement systems and will allow licensees and inspectors to make the measurements traceable to national standards.

Sub-Task A: Process Tank Volume Measurements Standardization

The final testing on the automated tank calibration apparatus was completed. The final report on the development and testing of the apparatus was presented at the American Nuclear Society Topical Conference on Measurement Technology for Safeguards and Material Control, Kiawah Island, South Carolina, November 26-29, 1979 ("Automated Accountability Tank Calibration", by D. Cooper, P. Baumgarten and B. Robertson). The apparatus has been moved to the scale house at NBS in preparation for demonstration of its use (in the first quarter of FY 81) in calibrating a 3000-liter tank which had previously been calibrated using volumetric test measures.

A paper, "In-Tank Measurement of Solution Density", by F. E. Jones, R. M. Schoonover and J. F. Houser, was presented at the Kiawah Island meeting. A modified version of the paper appeared in J. Res. Nat. Bur. Stand. (U.S.) 85, No. 3, May-June 1980 (219-221).

Two papers are undergoing editorial review. These relate to volume calibration of tanks using bubbler tubes. The titles for the two papers are: (1) Time Dependence of Pressure in a Bubbler Tube; and (2) The Dependence of Pressure in a Bubbler Tube on Liquid Properties. A. Gaigalas and B. Robertson are the authors of the first paper, and A. Gaigalas is the author of the second.

Sub-Task B: UF_6 Mass Measurement Implementation Program

The objective of this Sub-Task is the improvement of in-facility weighing of large UF_6 cylinders. The effort involves the implementation of a Measurement Assurance Program (MAP) for the measurement of the mass of UF_6 cylinders, filled with UF_6 or empty. The MAP provides continually verifiable evidence of performance of the mass measurement process, giving timely and quantitative uncertainty statements for the mass measurements.

A draft Standard Operating Procedure was sent to participating organizations for comment.

The 30B Replica Mass Standard (RMS) was first shipped to Oak Ridge to be used in calibrating the Oak Ridge In-House Standards. The RMS cylinder was then shipped from Oak Ridge to the General Electric Co. (G.E.) facility at Wilmington, NC, where it was used to recalibrate the G.E. In-House Standards, and then on to the Combustion Engineering Co. facility at Hematite, MO, and used to calibrate In-House Standards. The measurement data were sent to NBS for analysis.

The investigators for this Task are J. R. Whetstone, D. G. Cooper, G. P. Baumgarten, B. Robertson, A. K. Gaigalas, J. F. Houser, and E. Johnsen.

TASK IV: APPLIED MATHEMATICS FOR NUCLEAR SAFEGUARDS

This Task applies mathematics and statistics to the nuclear safeguards mission of NRC. The work in this Task consists of the preparation and publication of reports in five specific areas (Sub-Task A), and statistical support for other nuclear safeguards Tasks (Sub-Task B).

Sub-Task A: Statistical Methods in Nuclear Material Accounting

Assessing Errors Related to Characteristics of the Items Measured. A paper, "Assessing Errors Related to Characteristics of the Items Measured", by W. S. Liggett, appeared in Nuclear Materials Management IX, No. 1, 78-82 (1980). This paper (Report #1 of this Sub-Task) presents some problems in safeguards measurements that require further research. The abstract for the paper follows:

> Errors that are related to some intrinsic property of the items measured are often encountered in nuclear material accounting. An example is the error in nondestructive assay (NDA) measurements caused by uncorrected matrix effects. Such errors cannot be assessed by remeasurement of the items, and they cannot be fully assessed by measuring standards, although standards that span the range of the item characteristics might give upper and lower bounds. Nuclear material accounting requires for each material type one measurement method for which bounds on these errors can be determined. If such a method is available, a second method might be used to reduce costs or to improve precision. If the second method is less expensive than the first, then cost might be reduced by substituting the second method for the first in the measurement of some items. If the measurement error for the first method is longer-tailed than Gaussian, then precision might be improved by measuring all items by both methods.

Some Effective Statistical Approaches for Presenting Interlaboratory NDA Enrichment Measurements. In order to realistically evaluate measurements on prototype and primary standards measured by several different nondestructive assay (NDA) labs, it is necessary to distinguish real differences between laboratory findings and artificial differences, such as those caused by artifacts of peak area calculations. The first draft of Report #2 of this Sub-Task, "Empirically Calculated Weights for Finding Peak Areas in Gamma Spectroscopy",

by C. Spiegelman, provides guidance for locating and assessing the magnitude of these peak area artifacts. In particular, it provides a variety of algorithms for linear peak area computations which make the standards appear either most or least homogeneous under a variety of statistical criteria. The criteria include the usual F statistics, as well as the standardized range.

Robust Inference for Items Measured a Few Times. Estimation methods that de-emphasize the measurements that appear to be most in error offer the possibility of improved performance. This possibility, which occurs when the measurement error is non-Gaussian, is explored for some measurement designs not covered by previous work on robust methods. In these designs, each item in a collection of somewhat dissimilar items is to be measured two or three times, and the total for the entire collection is to be estimated. The methods are based on the assumption that the measurement errors for different items are from the same population. A method is proposed for each of three cases: three replicate measurements on each item, two replicate measurements by one method and one measurement by another method on each item, and two replicate measurements on each item. All the methods use the differences between the measurements on the same item to obtain a scale estimate by which the size of the measurement errors is judged. How the methods adjust the measurements differs from case to case. When there are three measurements per item, the most dissimilar of the three is downweighted. When there are only two measurements per item, the measurements for the entire collection are used to judge which measurements to downweight. The methods include estimates of the uncertainty due to measurement error, which are also obtained from the differences between the measurements on the same item. A report discussing these methods will be Report #3 of this Sub-Task.

The following is a preliminary abstract of Report #4 of this Sub-Task, which is in preparation:

Inventory Difference Calculations: Problems and Possibilities. The origin, development, and termination of the "error study" task funded by the Nuclear Regulatory Commission at the National Bureau of Standards is presented in this report. Other reports which grew (at least partially) out of this study are listed and related to the intent of this study.

A general approach to the calculation of Inventory Difference (ID) and its Limit of Error (LEID) was devised and refined through visits to a selected fuel-fabrication facility. This approach is discussed, with emphasis on the importance of having measurements "under control" and on the possibility of automating the computations. The concepts of "measurement process" and "Measurement Assurance Program" are also discussed and their importance shown. Possible problem areas, both those observed at the studied facility and others, are given, along with possible directions for resolution. Finally, new approaches to the calculation of ID and LEID being developed elsehwere are described and their relation to the approach being considered herein elucidated.

The following is the abstract of a report which is available as NBSIR 80-2151, and which has been submitted for publication in Technometrics:

A New Method of Assigning Uncertainty in Volume Calibration. This paper presents a practical statistical overview of the pressure-volume calibration curve for large nuclear materials processing tanks. It explains the appropriateness of applying splines (piecewise polynomials) to this curve, and it presents an overview of the associated statistical uncertainties. In order to implement these procedures, a practical and portable FORTRAN IV program is provided, along

with its users' manual. Finally, the recommended procedure is demonstrated on actual tank data collected by NBS.

The work with ASTM C26.06, the subcommittee on statistics of the committee on the Nuclear Fuel Cycle, included assumption of the chairmanship of the subcommittee by an NBS staff member. Two meetings were held to revise the draft standard on sampling nuclear materials in preparation for the July subcommittee meeting. At a meeting at Kiawah Island, SC, November 28-30, contributions were made to the revision of ANSI Standard N-15.22-1975, Calibration Techniques for Calorimetric Assay. Also, one staff member participated in the November 1979 meeting of the SALE Program Executive Committee and the general meeting of the participants.

Sub-Task B: Statistical Support for Other NBS Safeguards Tasks

Consultation, including two trips to Geel, Belgium, and preparation of an experimental design for the mass spectrometer measurements, was provided in support of the U_3O_8 SRM.

Mass spectrometer measurements on the ^{233}U SRM were analyzed, and the results were presented for inclusion on the SRM certificate.

Analysis of the first set of data from the UF_6 cylinder Mass Measurement Assurance Program was accomplished.

Investigators for this Task are W. S. Liggett, J. A. Lechner, and C. Spiegelman.

TASK V: USERS GUIDE, FACILITY ENGINEERING FOR SAFEGUARDS MEASUREMENT INSTRUMENTS

The primary objective of the Users Guide is to provide specific recommendations on how facilities should be designed to accommodate the necessary measurements to meet the SNM accountability goals of the safeguards program. The Guide will discuss how the measurement instruments and accountability systems should be installed and will outline the supporting services required. The Guide will also discuss facility design considerations so that the instruments can be efficiently calibrated, operated and repaired, either manually or remotely.

The preliminary draft of the Guide, which was completed during the previous fiscal year, was revised to reflect certain directives from the NRC. Discussions were held with the persons at Mound Laboratories who are preparing a Handbook of Nuclear Safeguards Measurement Methods. The purpose of these discussions was to assure that the two documents would be complementary.

The first rough draft of the Guide was completed and copies sent to contributors for comment and/or correction. In its draft form, the document had a length of approximately 18,000 words.

After the cut-off date for comments, May 30, 1980, the Guide was rewritten into its final form. It is now undergoing editorial review.

The investigator for this Task is E. Johnsen.

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Safeguards Standards Branch, Office of Standards Development
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10. SUPPLEMENTARY NOTES

☐ Document describes a computer program; SF-185, FIPS Software Summary, is attached.

11. ABSTRACT (A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here)

This report is a review of the progress made during Fiscal Year 1980 (October 1, 1979, through September 30, 1980) of a long-term NBS program sponsored by the Nuclear Regulatory Commission (NRC) to upgrade national measurements and standards capability for nuclear materials safeguards. This is the last report to NRC for this program, since NRC has terminated support to NBS as of September 30, 1980. The NRC-sponsored program at NBS was a synergistic part of the overall NBS program in this area which is sponsored jointly by the NRC, the Department of Energy (DOE), and the Department of State through the International Safeguards Project Office (ISPO). A summary of the progress for each of the five NRC-supported Tasks in the project is given. The first Task is concerned with the standardization of Non-Destructive Assay (NDA) methodology. The second Task involves the standardization of analytical chemical methodology. The third Task involves the standardization of bulk measurement technology. The fourth Task applies mathematics and statistics to nuclear safeguards. The fifth Task involves the development of a Users Guide to provide specific recommendations on how facilities should be designed to accommodate the necessary measurements to meet the SNM accountability goals of the safeguards program.

12. KEY WORDS (Six to twelve entries; alphabetical order; capitalize only proper names; and separate key words by semicolons)

analytical chemistry; annual report; bulk measurements; facility measurements guide; non-destructive assay; Nuclear Regulatory Commission; nuclear safeguards; standards; statistical support

13. AVAILABILITY

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